



Supplement of

Electrospray surface-enhanced Raman spectroscopy (ES-SERS) for probing surface chemical compositions of atmospherically relevant particles

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Figure S1. (a) Normal and (c, e) enhanced spectra of AS particles which were used for quantification of $I_{\text{SERS}}/I_{\text{NR}}$. (b, d, f) Magnified views of spectra at 950-1000 cm⁻¹ corresponding to $v(\text{SO}_4^2)$. Note that in the presence of aggregated Ag nanoparticles, enhanced spectra (e) showed strong peaks of citrate at 2945, 1395, and 932 cm⁻¹ corresponding to v(C-H), v(COO) and v(C-COO), respectively.



Figure S2. (a) Normal (blue) and (b) enhanced (red) spectra of AS/SA particles which were used for quantification of I_{SERS}/I_{NR} . (c, d) Magnified views of the corresponding spectra at 850-1050 and 2800-3100 cm⁻¹ corresponding to $v(SO_4^{2-})$ and $\delta(OH\cdots O)$, and v(C-H), respectively.



Figure S3. (a) Normal (blue) and (b) enhanced (red) spectra of AS/sucrose particles which were used for quantification of I_{SERS}/I_{NR} . (c, d) Magnified views of the corresponding spectra at 850-1050 and 2800-3100 cm⁻¹ corresponding to $v(SO_4^{2-})$ and v(C-H), respectively.



Figure S4. Plots of spray current versus applied voltage upon increasing and decreasing in the voltage.



Figure S5. UV-vis spectra of Ag nanoparticle suspension before (blue solid line) and after (red dash line) dilution with ethanol at 1:1 volume ratio.



Figure S6. Electrically mobility size distribution of Ag nanoparticle aerosols in the gas phase measured by DMA + CPC. The solid line was fitted to normal distribution.



Figure S7. (a, c) SEM images of and (b, d) size distributions of Ag nanoparticles after naturally dried suspension and deposited (electrosprayed) on the substrate, respectively. Inset images shows typical SEM images. 71 and 208 particles were totally counted to obtain the size distributions for (b) and (d), respectively. The solid lines were fitted to normal distribution.